

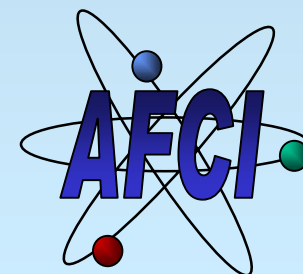


Development of Separations Technologies under the Advanced Fuel Cycle Initiative

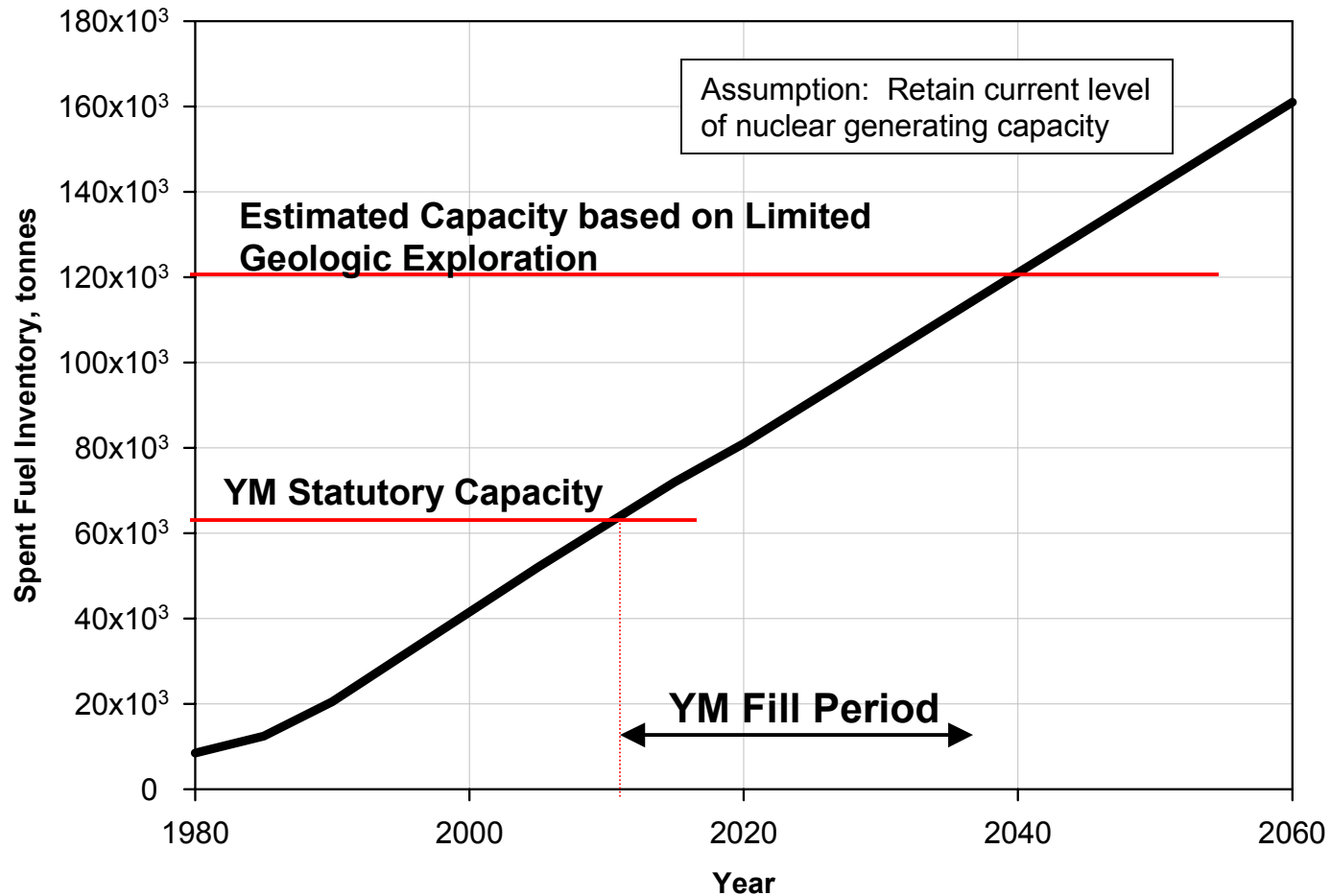
Dr. James Laidler
AFCI National Technical Director for
Separations Technology Development

Chemical Technology Division
Argonne National Laboratory

AFCI Semi-Annual Meeting
Albuquerque, New Mexico
January 22, 2003



Spent Fuel Generation in the U.S.



Composition of Spent Nuclear Fuel

Contents of 1 tonne PWR fuel (~ 2 fuel assemblies) at 50 MWd/kg burnup after cooling for 10 years:

955.4 kg U
8.5 kg Pu (5.1 kg ^{239}Pu)
0.5 kg ^{237}Np
1.6 kg Am
0.02 kg Cm
34.8 kg fission products

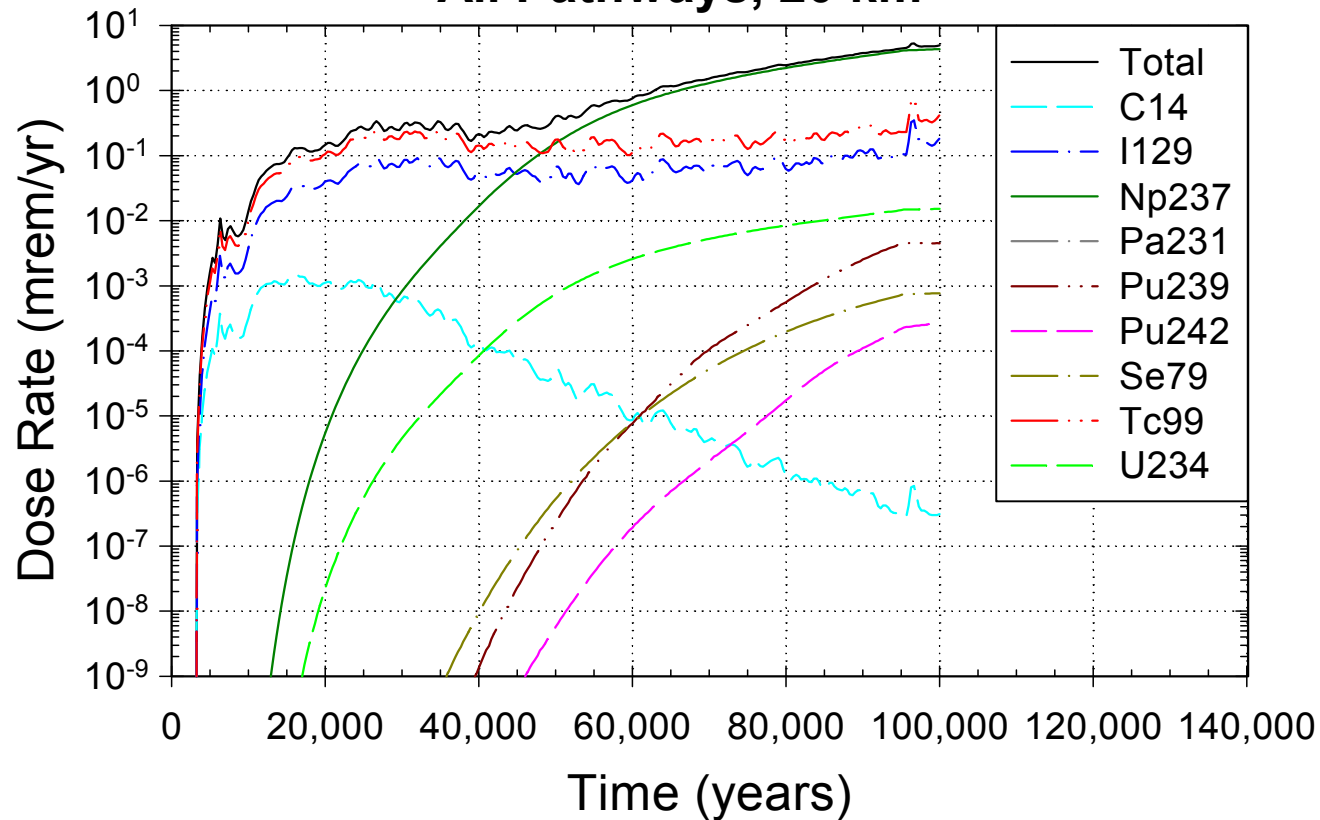
Fission Products:

10.1 kg Lanthanides
1.5 kg ^{137}Cs
0.7 kg ^{90}Sr
0.2 kg ^{129}I
0.8 kg ^{99}Tc
0.006 kg ^{79}Se
0.3 kg ^{135}Cs
3.4 kg Mo isotopes
2.2 kg Ru isotopes
0.4 kg Rh isotopes
1.4 kg Pd isotopes



Incentive for Removal of Certain Nuclides

TSPA-VA Base Case
100,000-yr Expected-Value Total Dose Rate History
All Pathways, 20 km



Principal Contributors to the Radiotoxicity of PWR Spent Fuel

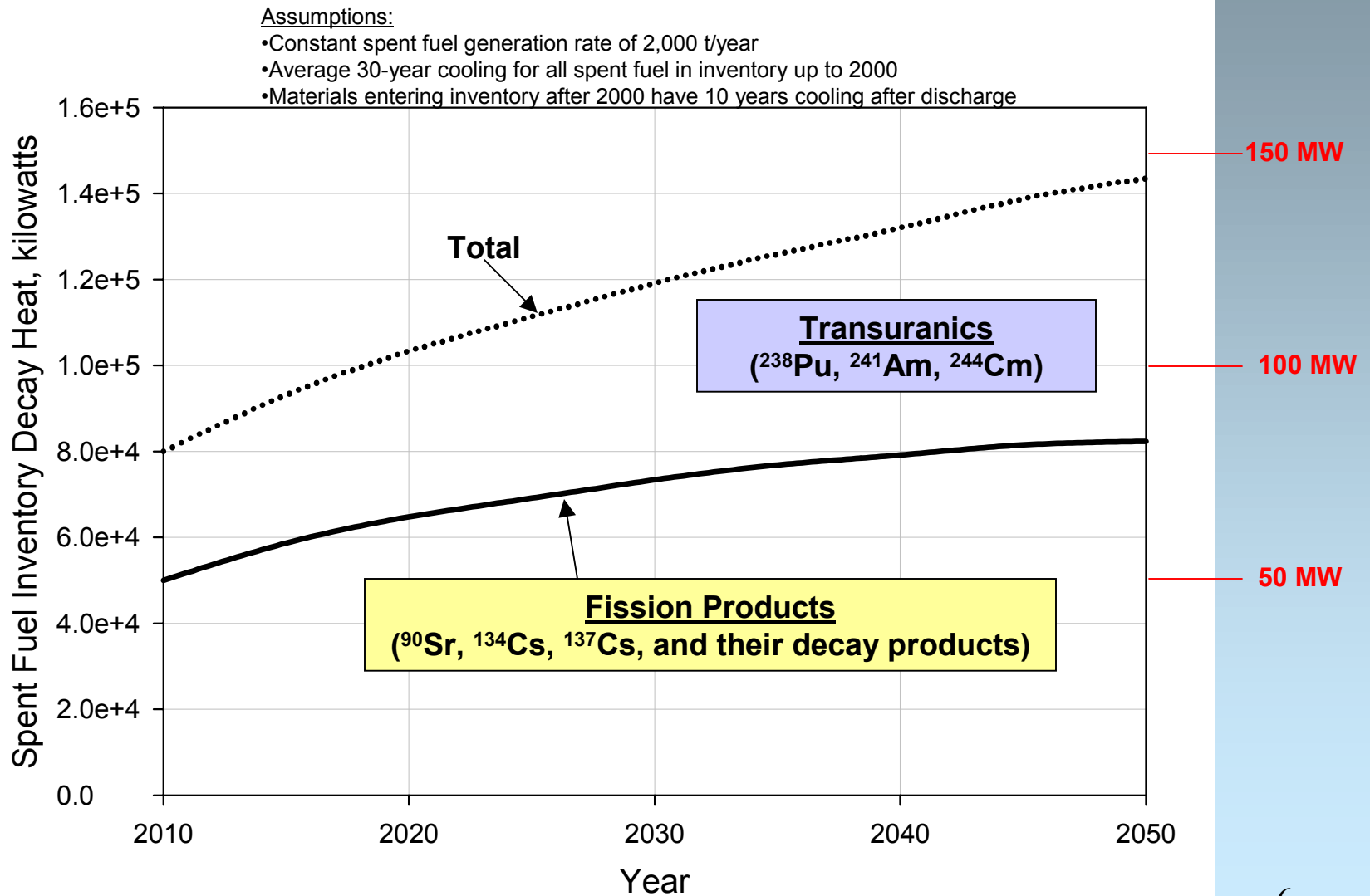
(50 MWd/kg burnup, 10 years' cooling)

<i>Isotope</i>	<i>Sv/tonne</i>	<i>Isotope</i>	<i>Sv/tonne</i>	<i>Isotope</i>	<i>Sv/tonne</i>
U-236	6.0E+02	Sr-90	9.2E+07	Y-90	8.9E+06
U-238	5.0E+02	Cs-134	1.4E+07	Ce-144	3.7E+04
Np-237	3.0E+03	Cs-137	6.3E+07	Pr-144	3.5E+02
Pu-238	3.5E+07			Pm-147	6.6E+04
Pu-239	2.8E+06			Sm-151	5.0E+03
Pu-241	2.0E+07			Eu-154	8.7E+05
Pu-242	2.0E+04			Eu-155	1.5E+04
Am-241	1.9E+07				
Am-243	7.7E+05			Ru-106	2.0E+05
Cm-244	4.9E+07				



(1 Sievert = 100 Rem)

Contributors to Heat Load



Role of AFCI

- Ameliorate spent nuclear fuel disposal issues
 - Greatly extend the time at which a second repository is needed; enable growth in nuclear generating capacity
 - Reduce the volume and heat load of high-level nuclear waste
 - Avoid the substantial costs associated with multiple future repositories
 - Reduce the risk of release of hazardous radionuclides to the environment
 - Place nuclear wastes in more durable forms; reduce release rates by orders of magnitude
 - Significantly reduce the radiotoxicity of materials sent to the geologic repository
- Facilitate the development of advanced reactor systems with closed fuel cycles and favorable economics
- Reduce inventories of civil plutonium; no future “plutonium mines”

Program Elements - Separations

- Series One Separations Technology
 - Treatment of spent nuclear fuel from current fleet of commercial light water reactors
- Series Two Separations Technology
 - Treatment of spent fuel from Gen IV reactor(s)
 - Recycle of minor actinide transmuter fuel/targets
- EBR-II Spent Fuel Treatment
 - Conditioning of EBR-II spent fuel and blankets to comply with agreement with State of Idaho for removal by 2035
 - Development of advanced process equipment and methods for spent fuel treatment in production-scale plants



Support of AFCI Major Goals

- Reduce the cost of geologic disposal of high-level nuclear waste
 - Reduce annual waste volume generation rate from 2,210 m³ per year to about 110 m³ per year by removing uranium, transuranics, and heat-generating fission products
 - Reduce high-level waste mass from 2,600 tonnes per year to about 600 tonnes per year (most of this is cladding hulls and assembly hardware)
 - Reduces number of waste packages and drip shields
 - Reduces facility requirements for forced ventilation
 - Reduces heat load by 97-99%
- Reduce fuel cycle costs
 - Reduce spent fuel treatment costs to levels near 1 mill/kW_e-h (~\$400/kg)



Support of AFCI Major Goals (cont.)

- Reduce inventories of civil plutonium
 - Amount of plutonium sent to repository disposal will be reduced from about 17,000 kg per year to less than 75 kg per year
- Reduce toxicity of high-level nuclear waste
 - Radiotoxicity of high-level waste sent to geologic repository will decrease to less than that of the original uranium fuel in a storage time of less than 1,000 years*, well within the lifetime of current disposal containers
 - Radiotoxicity at time of disposal will be less than 1% of that of the comparable amount of unprocessed spent fuel
 - Long-lived fission products (^{99}Tc and ^{129}I) can be separated for transmutation or placed in durable waste forms that will reduce their dose risk by a factor of 100



* Assuming 99.5% overall recovery of transuranic elements

Series One Elements

- Advanced aqueous process development
- Pyrochemical process development (LWR fuel)
- Engineered product storage
- Spent fuel treatment facility design support



Advanced Aqueous Process Development

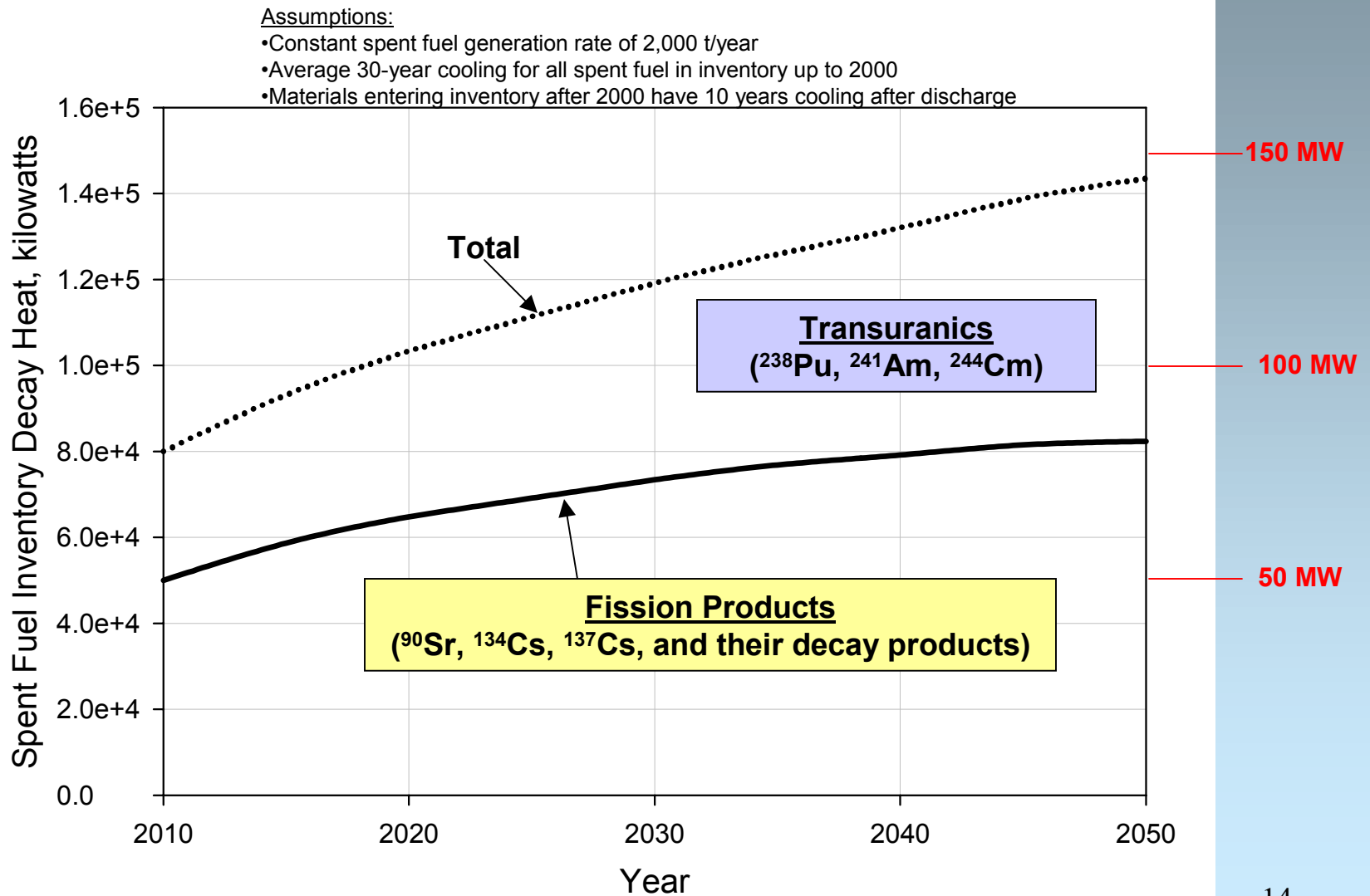
- Wrapup of UREX demonstration conducted in FY02
- UREX+ process development
 - AMUSE code development
 - Individual process operations development
- Process demonstrations
 - Small-scale tests with hot fuel
 - Large-scale engineering demonstration (up to 20 t/y scale)



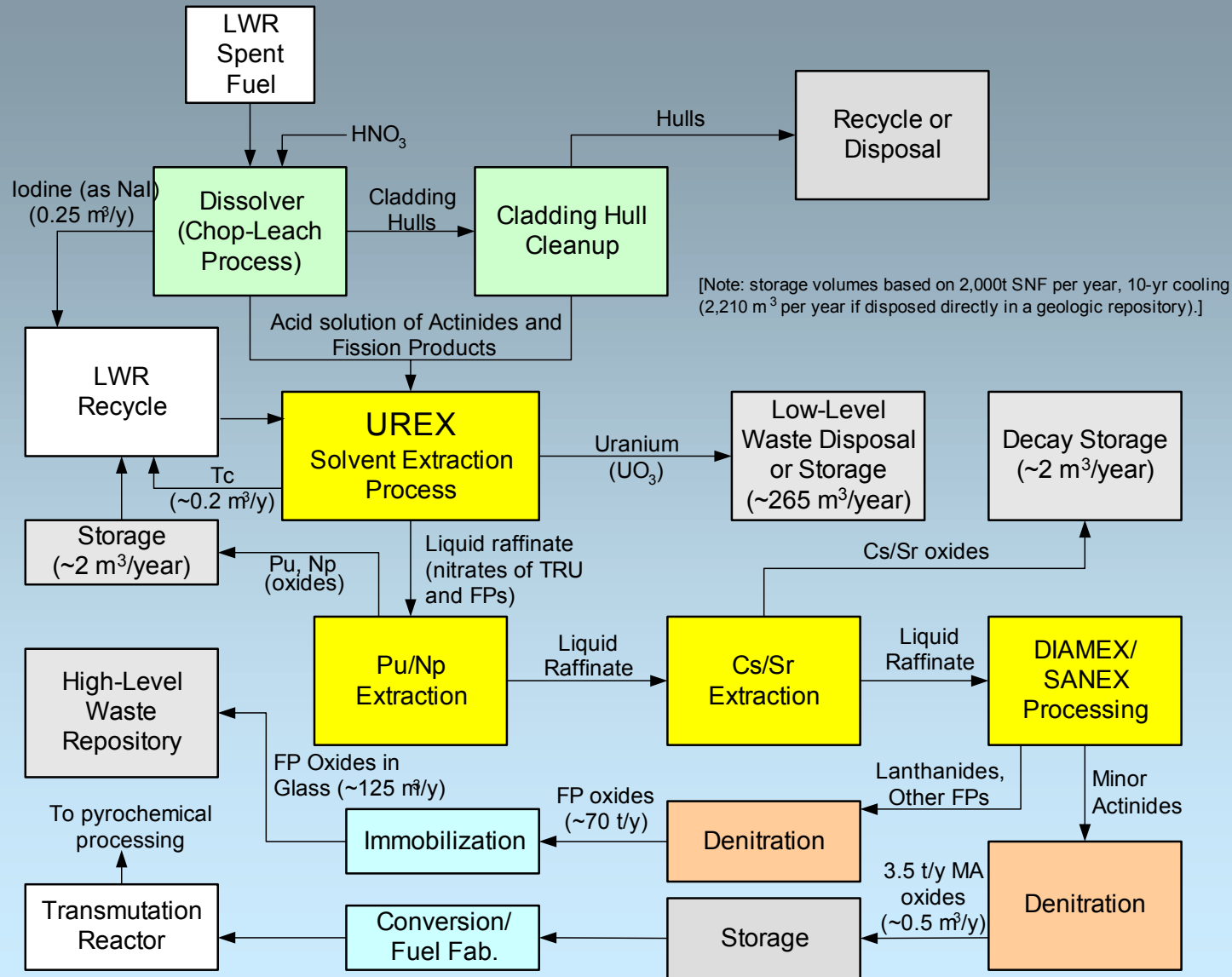
Targeted Radionuclides

- Offsite dose risk
 - ^{99}Tc , ^{129}I , $^{237}\text{Np}/^{241}\text{Am}$
- Radiotoxicity
 - ^{238}Pu , ^{239}Pu , ^{241}Pu , ^{241}Am , ^{244}Cm
 - ^{90}Sr , ^{134}Cs , ^{137}Cs , ^{90}Y
- Heat Generation
 - ^{238}Pu , ^{241}Am , ^{244}Cm
 - ^{90}Sr , ^{134}Cs , ^{137}Cs (and Ba/Y decay products)

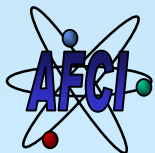
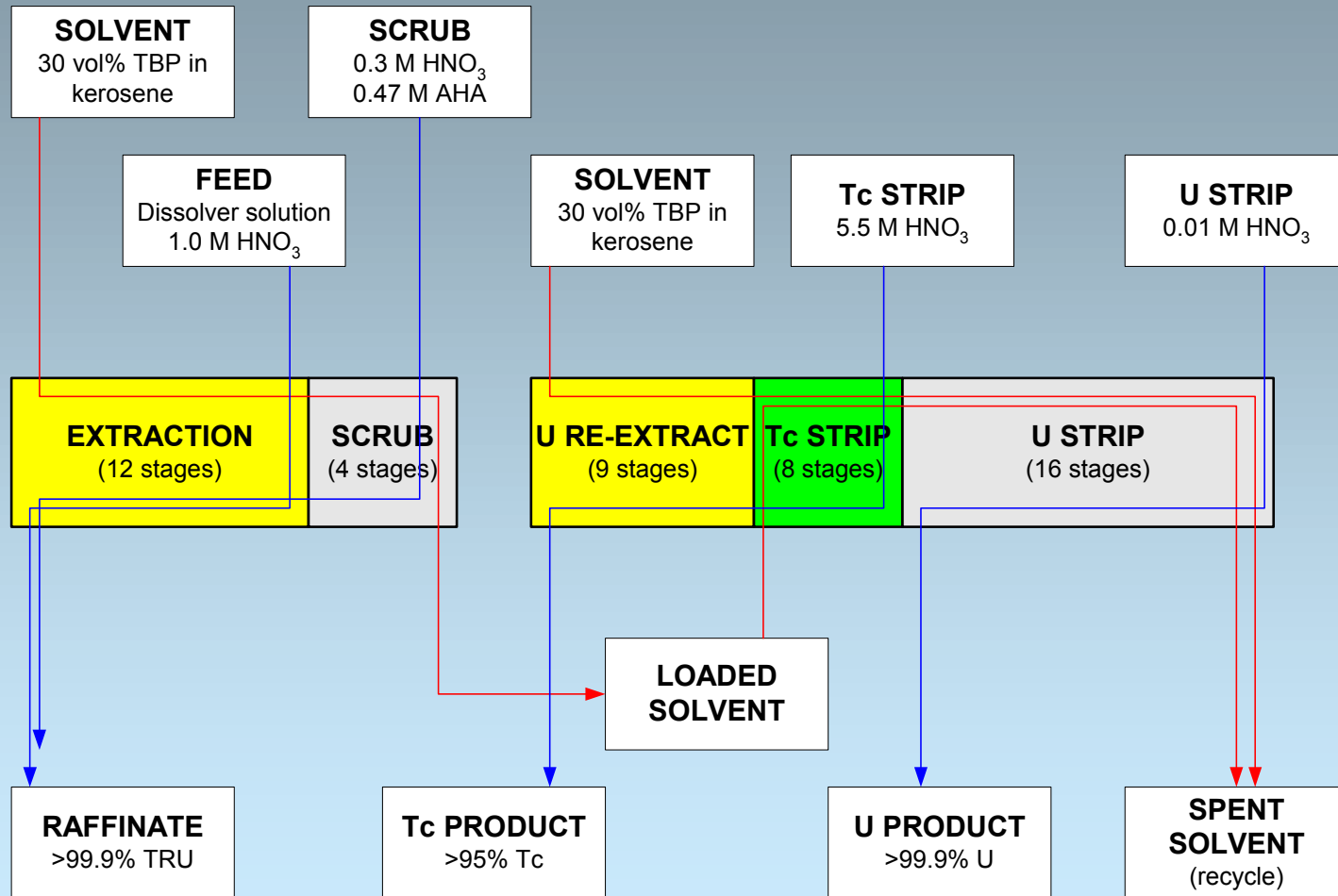
Contributors to Heat Load



UREX+ Process for LWR Spent Fuel



UREX Process Flowsheet

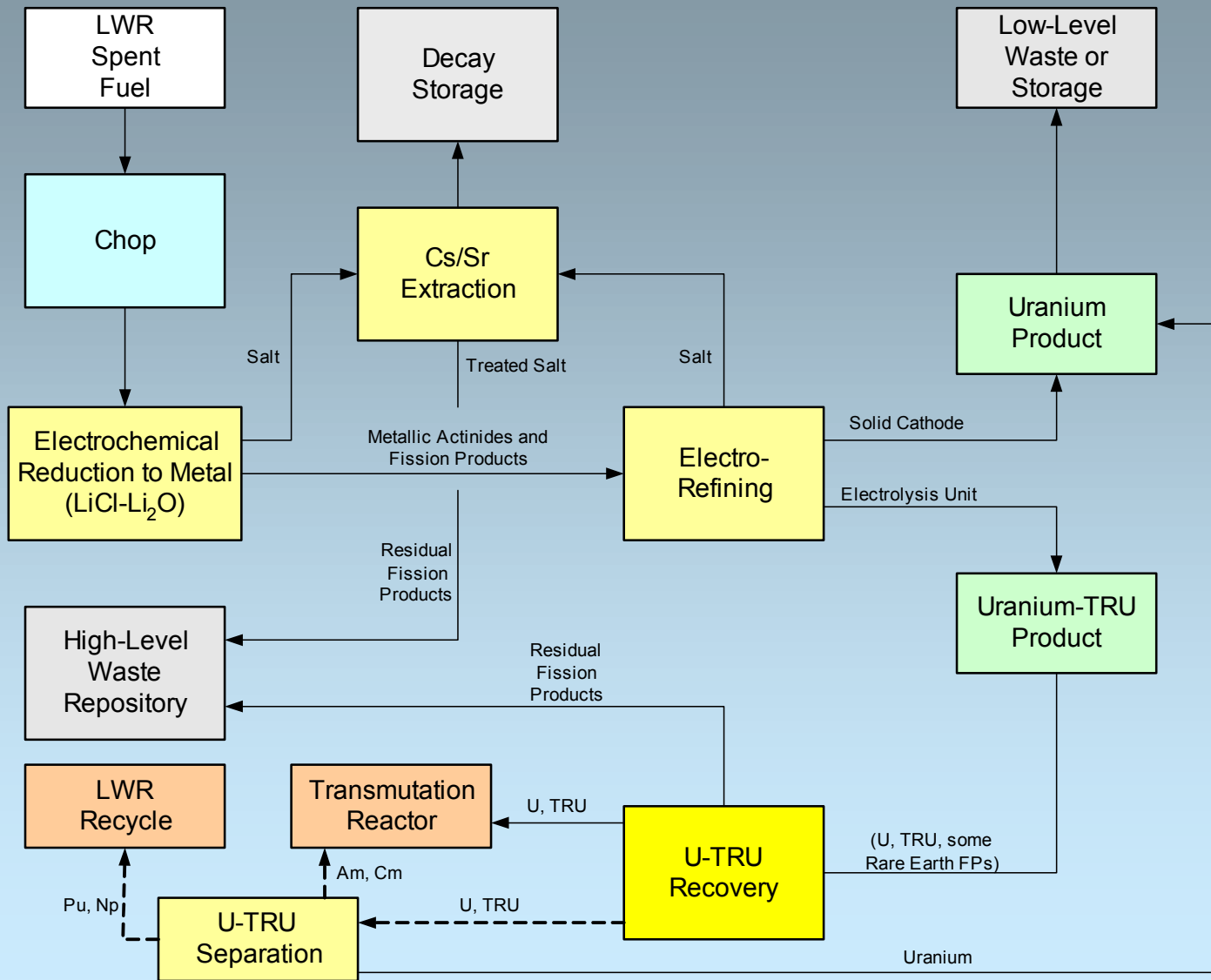


Pyrochemical Process Development

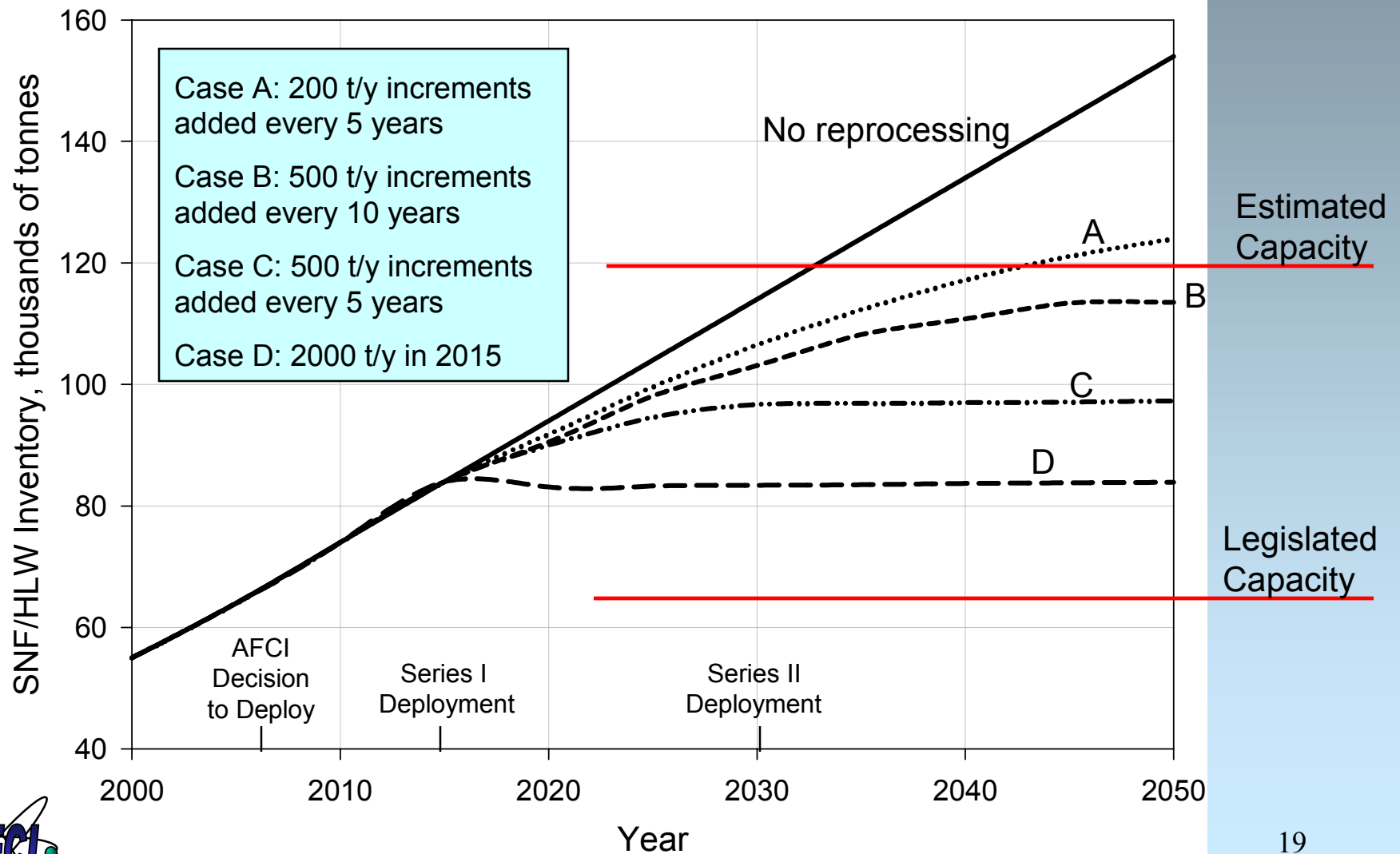
- PYROX process development
 - Application to LWR oxide fuel
 - Best use is in direct recycle to fast reactors
 - Requires hybrid process for thermal recycle of Pu (Np)
- Hybrid process development
 - Combines with UREX+ to produce TRU product for fast reactor recycle (PYRO-A process)
 - Can be used to separate TRU from lanthanides (rare earth fission products) for Series One recycle



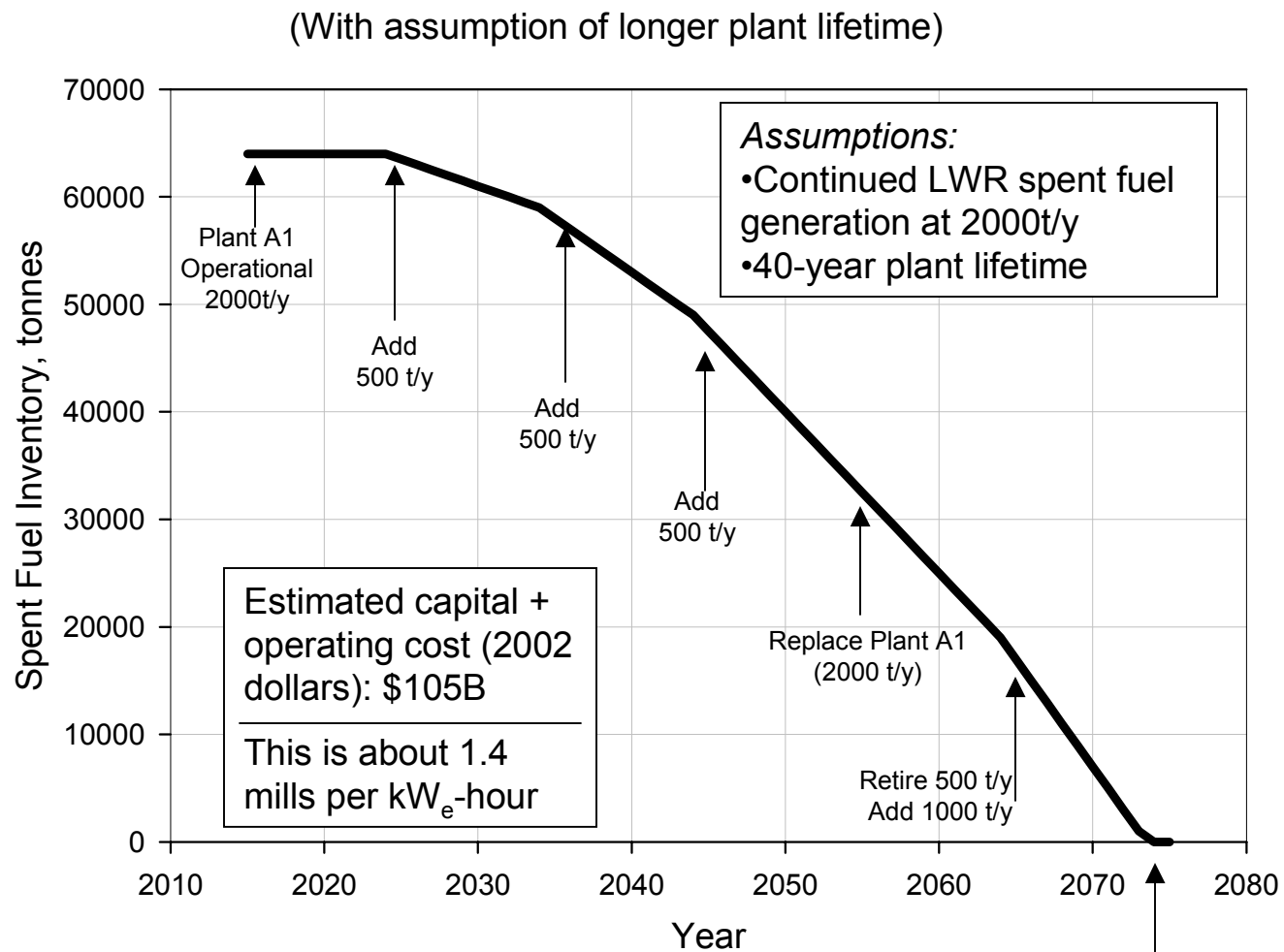
PYROX Process for LWR Spent Fuel



Effects on Repository Capacity



Possible Scenario to Eliminate Backlog



Eliminates SNF inventory and provides 3500 t/y capacity to support nuclear system



Highlights: Series One Separations

- Process down-selection by the end of FY2007
- Small-scale demonstration of UREX+ process in FY2003-FY2004 to validate and optimize the flowsheet
- Preparation for engineering-scale UREX+ demo at INEEL
 - Major efforts in FY2004-FY2005
 - Demonstration starts no later than FY2007, may continue beyond to provide Pu for Series One fuel LTA irradiations
- PYROX process demonstrations carried out at similar scale
 - Initial test in FY2003, 20-50kg batch test to be completed in FY2006-FY2007



Highlights: Series One Separations (cont.)

- Engineered Product Storage task directed toward the handling of the various product streams from the separations process (U, Pu/Np, Cs/Sr, Am/Cm)
 - Mainly paper studies initially, then limited experimentation
 - Definition of preferred storage forms by 09/06
- Spent Fuel Treatment Facility Design Support task
 - Deployment strategy options (size, timing, siting)
 - Separations process criteria (essential to process down-selection) [interim: 09/03; final: 09/04]
 - Development of Functional and Operational Requirements [interim: 09/03; final 09/04]
 - Pre-conceptual design study for large plant (more later)
 - Existing structures
 - Green field plant
 - Technology-neutral; aqueous or pyro until down-selection in 2007 or earlier



What Constitutes an Adequate Demonstration?

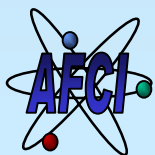
- Investment in the 2015 large spent fuel treatment facility will be very large; need solid assurance that the process to be installed in it will be successful.
- UREX+ process chemistry can be proven at very small scale with hot fuel, but fluid hydraulics must be proven at a scalable size and process integration must be demonstrated.
- PYROX process, basically a batch process, must be proven in integrated form; the demonstration batch size can be rather small.
- The reliability of both processes must be demonstrated, by operation over extended periods of time with equipment failure or process upset

UREX+ Engineering-Scale Demonstration

- For demonstration with centrifugal contactors, throughput rate of 0.4 – 1.5 liter/minute is sufficient

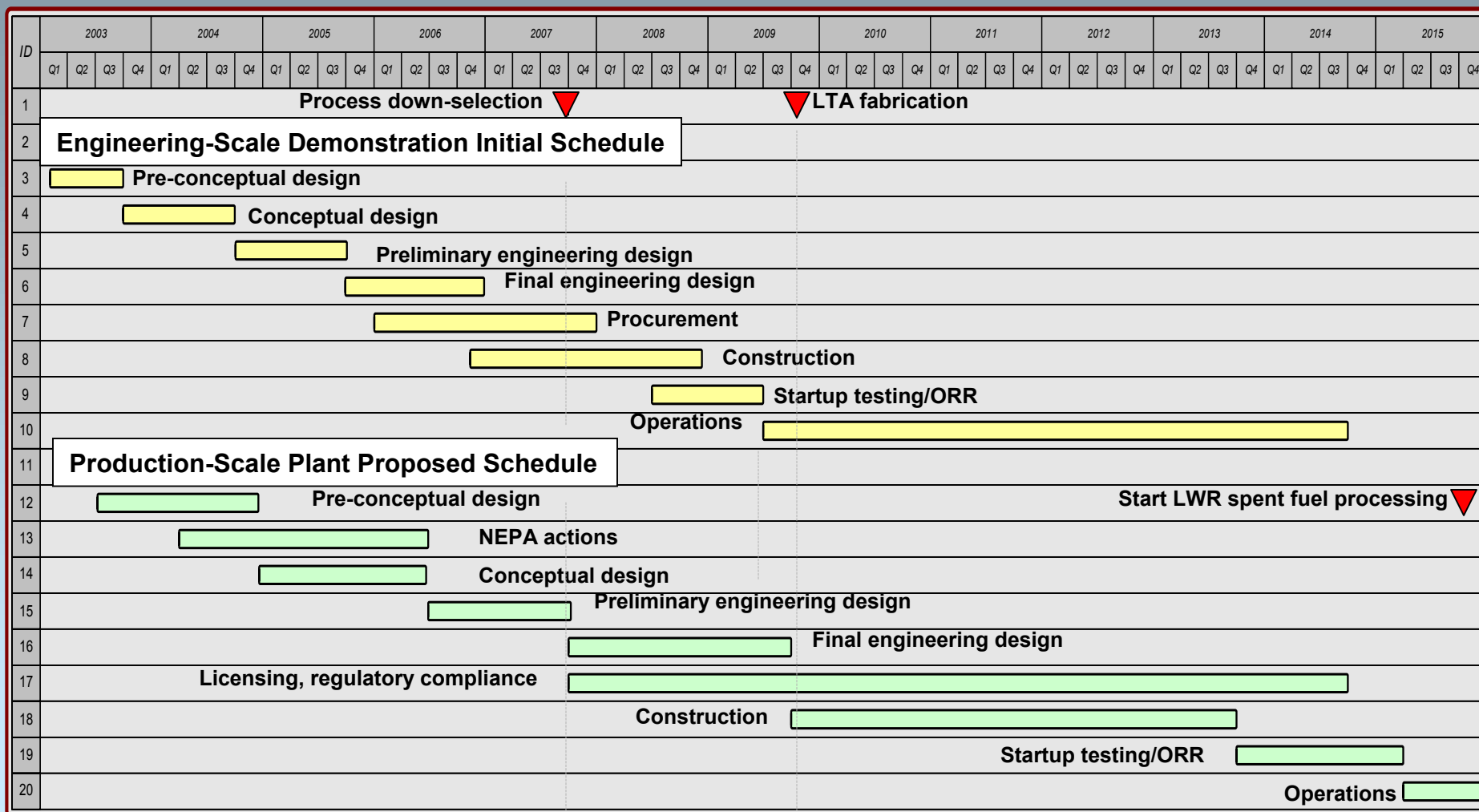
<i>Rotor dia.</i>	<i>Feed rate</i>	<i>SF per year</i>
4 cm	0.4 liter/min	7 tonnes
5.5 cm	1.5 liter/min	25 tonnes

- Corresponds to 0.35% -1.25% of production throughput
- Extended demonstration: five 30-day runs
- About 5 – 20 tonnes of spent fuel processed in demonstration
- Initial plan for engineering-scale demonstration: 30 tonnes total, to meet LTA fabrication needs (300 kg Pu-Np)


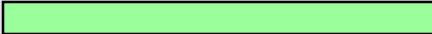





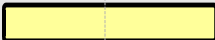




Separations Working Group Meeting 12/10/02 – Problem with Initial Demonstration Schedule

(Preliminary demonstration schedule as presented by INEEL)



Preferred Schedule for UREX+ Engineering-Scale Demonstration

ID	2003				2004				2005				2006				2007				2008				2009				2010							
	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4								
1																	 Process down-selection																			
2	Final design, large plant																																			
3					Pre-conceptual design																															
4									Conceptual design																											
5													Preliminary engineering design																							
6																	Final engineering design																			
7																	Procurement																			
8																	Construction																			
9																					Startup testing/ORR															
10													Operation																							

PYROX Engineering-Scale Demonstration

- PYROX comprises a series of batch processes
 - Oxide reduction
 - Uranium electrorefining
 - TRU recovery by electrolysis (may be semi-continuous)
 - Cathode processing
- Demonstration of each step is required; batch size of 20-50 kg spent fuel per day
 - Corresponds to ~4-10 tonnes spent fuel per year (0.2-0.5%)
 - Extended demonstration: five 30-day runs
- Process will not provide separated Pu-Np for LTA fabrication
 - Auxiliary steps would be required

ID	2003				2004				2005				2006				2007				2008				2009				2010			
	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4
1	<div> <div></div> <div>Process down-selection</div> </div>																															
2	<div> <div></div> <div>Final design, large plant</div> </div>																															
3	<div> <div></div> <div>Equipment Design</div> </div>																															
4	<div> <div></div> <div>Element chopper-shredder</div> </div>																															
5	<div> <div></div> <div>Oxide reduction</div> </div>																															
6	<div> <div></div> <div>Electrorefining</div> </div>																															
7	<div> <div></div> <div>TRU recovery</div> </div>																															
8	<div> <div></div> <div>Cathode processing</div> </div>																															
9	<div> <div></div> <div>Equipment Fabrication</div> </div>																															
10	<div> <div></div> <div>Element chopper-shredder</div> </div>																															
11	<div> <div></div> <div>Oxide reduction</div> </div>																															
12	<div> <div></div> <div>Electrorefining</div> </div>																															
13	<div> <div></div> <div>TRU recovery</div> </div>																															
14	<div> <div></div> <div>Cathode processing</div> </div>																															
15	<div> <div></div> <div>Process Demonstration</div> </div>																															
16	<div> <div></div> <div>Commercial fuel preparation</div> </div>																															
17	<div> <div></div> <div>Fuel shredding</div> </div>																															
18	<div> <div></div> <div>Oxide reduction</div> </div>																															
19	<div> <div></div> <div>Electrorefining</div> </div>																															
20	<div> <div></div> <div>TRU recovery</div> </div>																															
21	<div> <div></div> <div>Cathode processing</div> </div>																															

AFCI Separations Activity Teams

- UREX+ Process Design Team
 - Chair: G. Vandegrift (ANL)
 - Members: T. Todd (INEEL), G. Jarvinen (LANL), E. Collins (ORNL), T. Rudisill (WSRC)
- Separations Deployment Options Team
 - Chair: B. Boore (WSRC)
 - Members: D. Graziano (ANL), E. Collins (ORNL), R. Henry (INEEL), D. McGuire (WSRC)
- Separations Criteria Development Team
 - Chair: E. Collins (ORNL)
 - Members: G. Vandegrift (ANL), M. Williamson (ANL), M. Goff (ANL), G. Jarvinen (LANL), T. Todd (INEEL)
- F&OR Development Team
 - Chair: R. Henry (INEEL)
 - Members: G. Vandegrift (ANL), C. Lovejoy (LANL), E. Collins (ORNL), B. Boore (WSRC), M. Goff (ANL)
- Pre-conceptual Design Team
 - Chair: R. Henry (INEEL)
 - Members: B. Boore (WSRC), E. Collins (ORNL), K. Budlong-Sylvester (LANL), M. Goff (ANL), G. Jarvinen (LANL)
- Engineered Product Storage Team
 - Chair: D. Bennett (LANL)
 - Members: G. Vandegrift (ANL), E. Collins (ORNL), N. Schroeder (LANL), G. Kessinger (WSRC), W. Halsey (LLNL)

Series Two Separations

- Gen IV fuel treatment process development
 - Nitride fuel
 - Gas-cooled reactor fuel (once fuel design is established)
 - Very high temperature reactor (VHTR)
 - Gas-cooled fast reactor (GFR)
 - Metallic fuel
 - Other fuel types as appropriate
- Advanced Processing Concepts Development
 - Innovative process concepts that have the potential to reduce costs, simplify operations, and minimize wastes
 - Actinide crystallization process, advanced dissolvers, improved extraction methods, fuel shredder
 - Potential for increased funding requirements as practical innovations are identified



EBR-II Spent Fuel Treatment

- Pyroprocessing of EBR-II spent driver fuel and blankets mandated by ROD signed by W. Magwood as the preferred option for treatment of this material to meet State of Idaho requirements for removal of high-level wastes from the State by 2035
- Emphasis now on development of advanced technology concepts for production-scale operations with pyrochemical processes
 - Applicable to Series One and Series Two (viz, Gen IV)



Advanced Pyroprocessing Technology Concepts

- Electrolyzer for TRU recovery; replaces liquid cadmium cathode system
- Hybrid process for LWR spent fuel treatment; coarse separation of lanthanides from TRUs
- Development of high-capacity uranium electrorefiner; required for treatment of LWR spent fuel, also expedites blanket treatment
- Development of high-throughput product processing equipment; semi-continuous process for consolidation of U and U-TRU products
- Development of high-efficiency fuel shredder; supports both aqueous and pyrochemical processing, increases recovery efficiency for actinides



Pyroprocess Waste Forms

- Near-term effort on development of Waste Acceptance Product Specification, Waste Compliance Plan, and Waste Qualification Report for disposal of EMT wastes in Yucca Mountain
 - These waste forms are common to all Series Two treatment processes and may also be used in Series One processes
- Production-scale process equipment for waste form preparation is also being pursued

